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Fundamental insight in soot oxidation over a Ag/Co_3O_4 catalyst by means of Environmental TEM

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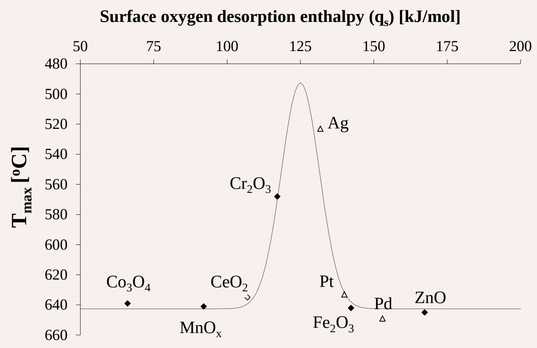
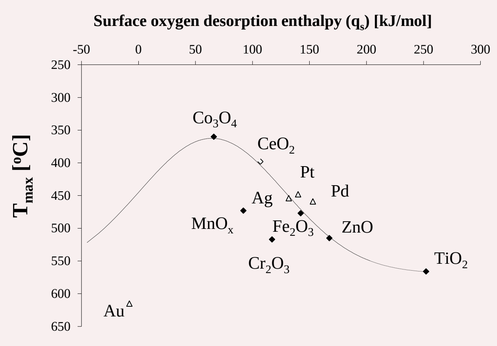
Motivation: **Soot emitted from diesel engine as typical solid particles has caused acute health problems to human beings. In modern vehicles, soot filters need to be regenerated periodically through a catalyzed high temperature oxidation process involving an extra fuel consumption. A dream catalyst would oxidize soot at a very low ignition temperature T_{ig}, ideally the temperature of the exhaust gas itself (T_{ig} < 250 ºC).**

Choice of the materials:

Soot oxidation is a gas/solid/solid interaction. When catalyst and soot are crushed together (tight contact) the oxidation occurs at lower temperature, than when the two are stirred together with a spatula (loose contact).





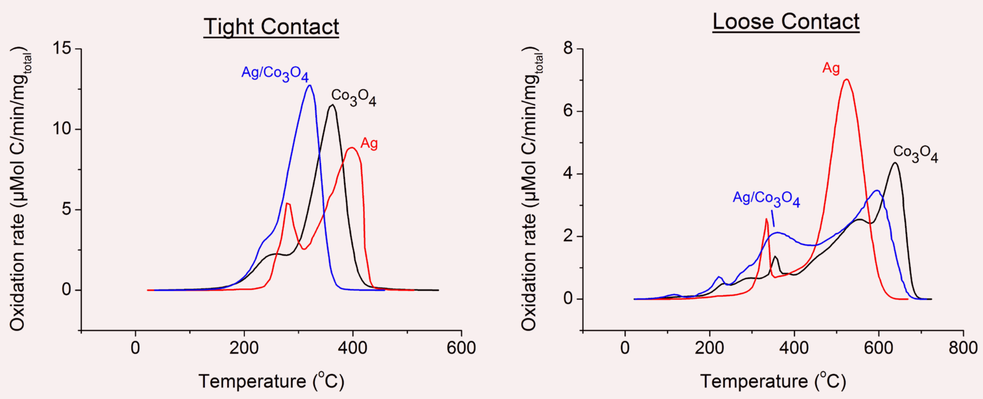
Catalytic test of Ag/Co_3O_4, Ag and Co_3O_4:

Temperature programmed oxidation (TPO) in a flow reactor setup.

10-18 mg soot/catalyst mixture (ratio: 1/5 Wt/Wt).

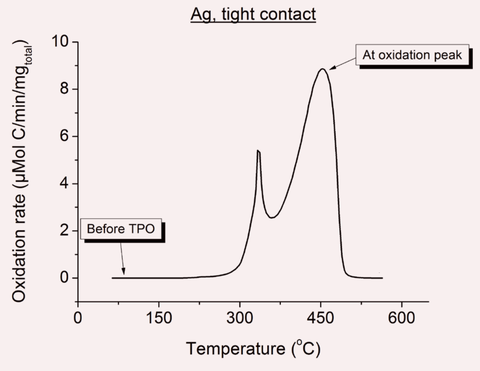
1 NL/min flow of 10 vol% O_2 in N_2. 100 to 750 ºC at a rate 11 ºC/min.

CO and CO_2 concentrations in the effluent gas are measured with an ABB AO2020 continuous IR gas analyzer.

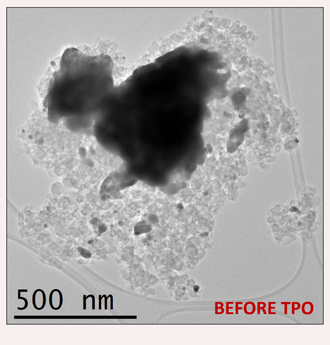


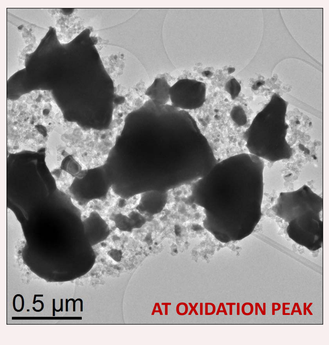
→ Performance of cosupported catalyst <u>cannot be directly described in terms of the activity of the single Ag and Co_3O_4 components</u>

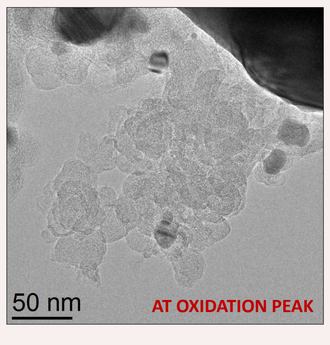
Ex situ TEM analysis: TPO snapshots of Ag/soot system.



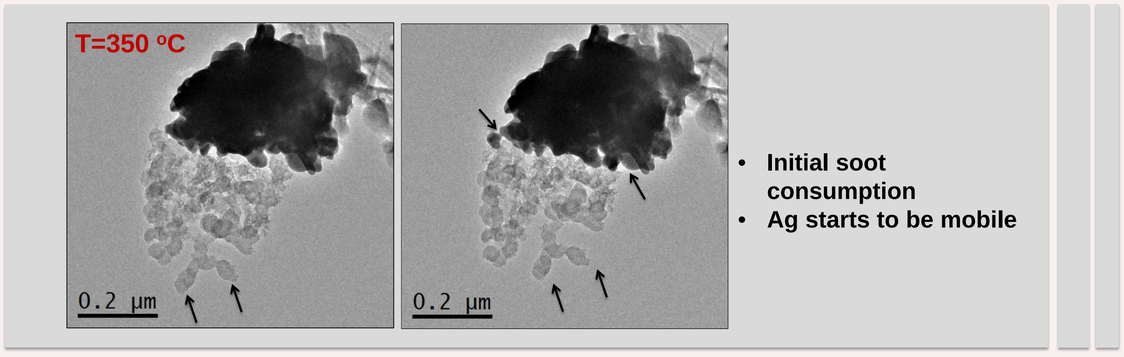
- **Tapering of agglomerate's borders at oxidation peak**
- **Soot consumption**



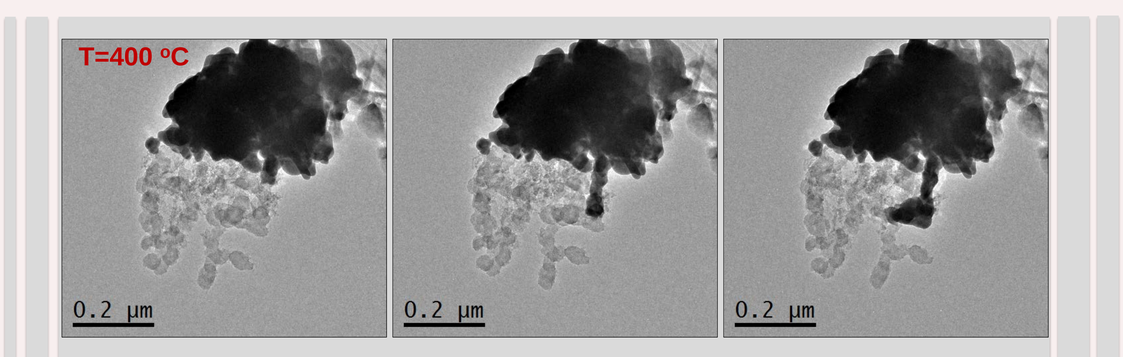


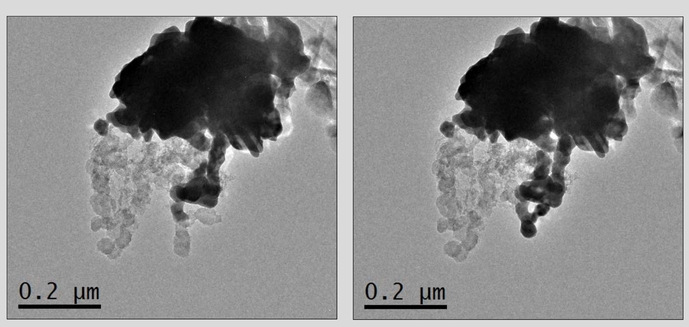


In situ TEM analysis:

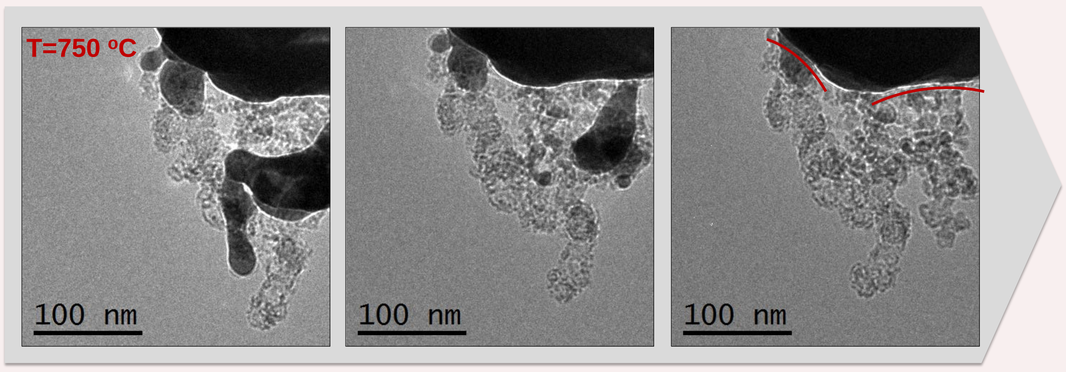


- **Initial soot consumption**
- **Ag starts to be mobile**





- **Highly mobile Ag**
- **Soot get consumed as Ag advances**
- **Diffraction halos are visible for the advancing Ag (crystallinity)**



→ Soot oxidation is an exothermic process. Local heating could lead to melting of the advancing Ag (seen on metal nanoparticles in literature [1] for graphite oxidation). Once oxidation is over, Ag could solidify again.

→ At higher temperature, Ag agglomerates back leaving a soot "snake skin" and forming the tapered fronts.

References:

[1] J. R. Fryer, Nature (1968) 220, 1121-1122

Aknowledgements:

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